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Mode-Locked Deceleration of Molecular Beams: Physics with Ultracold Molecules

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## Final Performance Report

"Mode-Locked Deceleration of Molecular Beams: Physics with Ultracold Molecules"

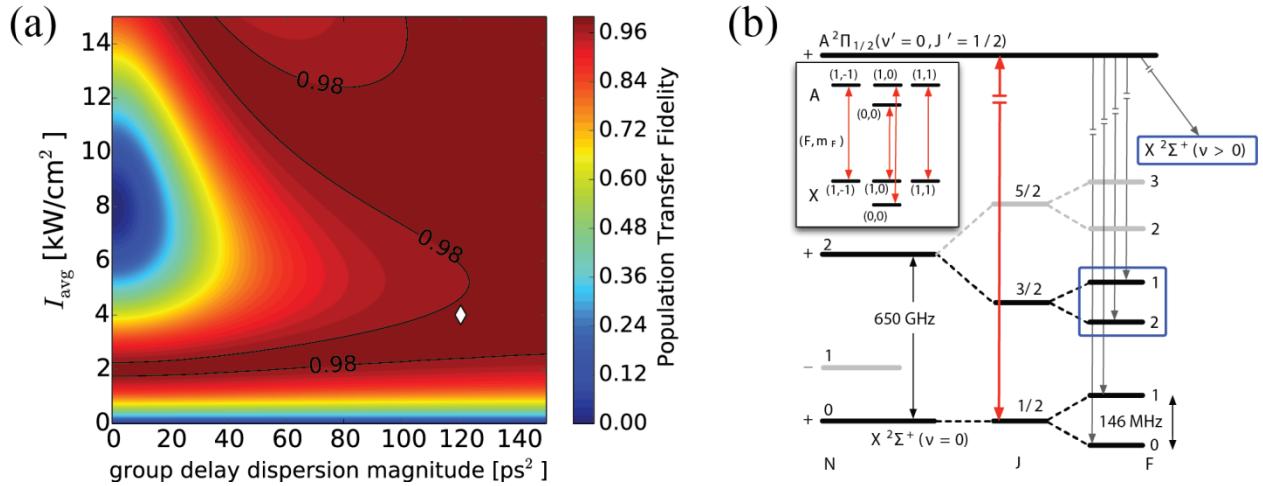
P.I. Wesley C. Campbell

Report Period: April 1, 2013- March 30, 2016

As a direct result of the support of this award, we constructed a MOT (Magneto Optical Trap) of rubidium atoms, demonstrated excitation of the laser-cooled atoms with single pulses from a mode-locked laser, constructed a buffer-gas beam source, developed a novel long-pulse autocorrelator for measuring and characterizing pulse chirp for adiabatic rapid passage, and demonstrated laser cooling and trapping of atoms on a two-photon transition driven by an optical frequency comb.

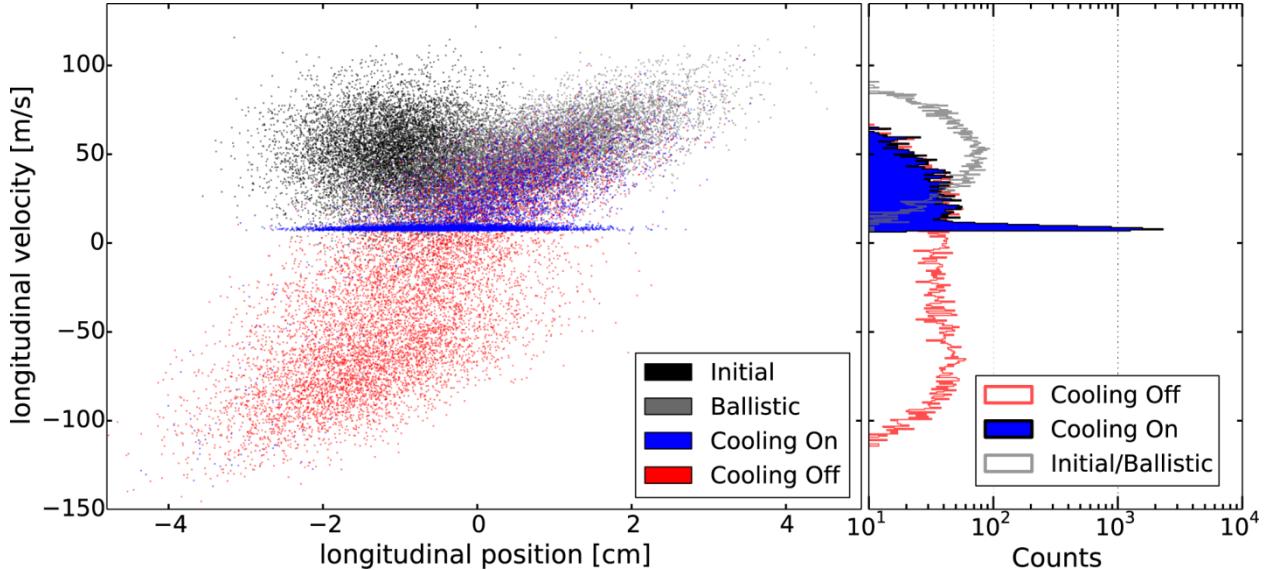
### Section I. Theoretical Studies

In order to guide our experimental efforts for using pulses from a mode-locked laser to decelerate a molecular beam, we undertook a theoretical study of the details of this process. The technique of Adiabatic Rapid Passage (ARP) was identified in our proposal as a way to boost the population transfer fidelity needed for momentum transfer to slow a molecular beam. We identified the relevant structure of the diatomic molecule SrH for driving this process as a demonstration and solved the Schrodinger equation numerically to obtain the population transfer fidelity as a function of the pulse energy and how much frequency chirp was added. Our model for adding frequency chirp is to send each pulse through a linear medium with a large amount of group delay dispersion (GDD), and the results are shown in Fig. 1(a). This calculation shows that the population transfer fidelity becomes insensitive to pulse energy, and we have tried to include all of the features of the experimental system that may affect this in the calculation, such as the multi-level structure of the molecules and polarization impurity. We calculate that the operating conditions shown can be achieved, though the creation of sufficiently large GDD remains to be demonstrated.



**Figure 1** (a) Population transfer fidelity calculated for a single pulse as a function of pulse energy (in units of time-averaged laser power) and the pulse chirp (calculated as the amount of GDD applied to a transform-limited pulse). This numerical solution to the Schrodinger equation uses the level structure shown in the inset in (b). Details can be found in the publication (ref. [1]).

While the deceleration calculation indicates that a molecular beam can be slowed, we also investigated a cooling technique known as single-photon cooling. The distinction between slowing and cooling that we make here is that deceleration can translate the velocity distribution without changing its width, while cooling removes entropy and can therefore compress the width. The appeal of using single photons to cool the molecules is its efficiency—since photons will carry away entropy via spontaneous emission and it is difficult to repeat spontaneous emissions many times with molecules, we wish to get as much cooling as possible with the fewest possible spontaneous emission events.



**Figure 2** Monte-Carlo simulations of mode-locked deceleration of a molecular beam with single-photon cooling. The black and gray distributions represent the molecules in the absence of any laser fields. The red points show the effect of the mode-locked deceleration only, while the blue curve has both the deceleration and the single-photon cooling lasers illuminating the atoms. For the latter, the density increases near the target velocity, and the velocity distribution shows clear cooling.

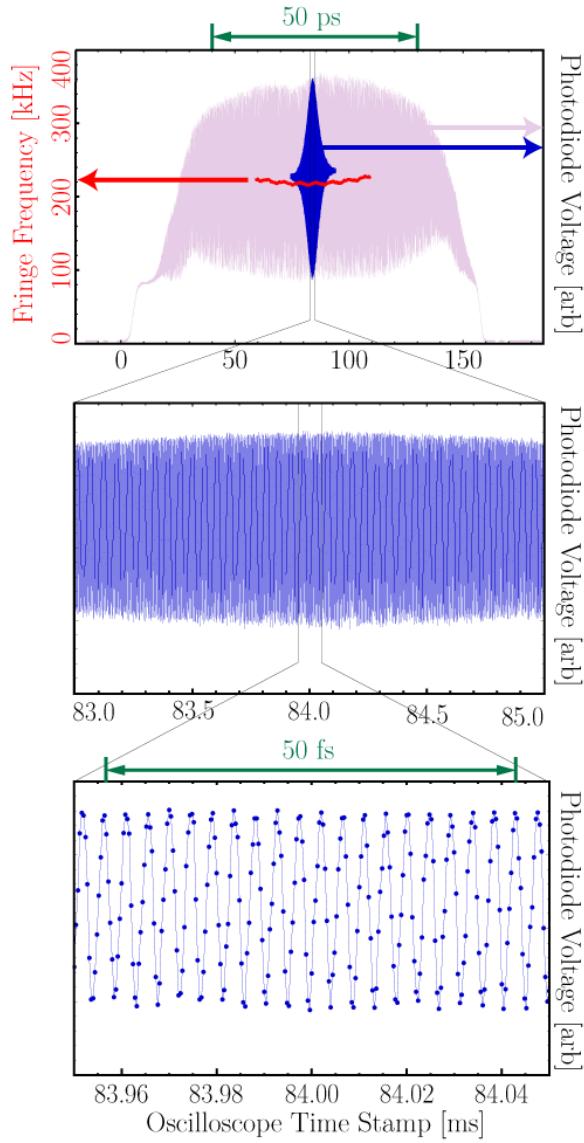
The single-photon cooling process we simulated is a cooling in velocity space, but a position-dependent version is also possible and works the same way. A continuous-wave (CW) laser illuminates

the molecular beam during its pulsed deceleration. The CW laser should be resonant with a transition that will optically pump the molecules into a dark state. Since the CW laser is narrow-band compared to the Doppler width of the beam, the only molecules that are optically pumped into the dark state are those with the desired target velocity. Every spontaneously-emitted photon that optically pumps molecules into the dark state therefore heralds the arrival of a molecule in the target velocity class, dramatically reducing its entropy and compressing phase space density. Monte-Carlo simulations of the pulsed deceleration with the single-photon cooling laser are presented in Fig. 2.

## Section II. Single pulses: Characterization and Atomic Excitation

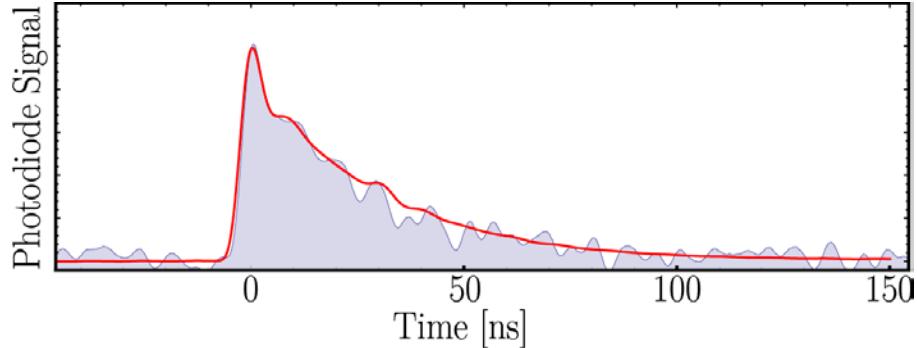
Our deceleration technique requires us to effect robust population transfer to decelerate the molecular beam using chirped pulses that may be tens of picoseconds long. Characterizing pulses of this length is arguably more challenging than sub-picosecond pulses, because a scanning autocorrelator must be able to produce delays that cannot be achieved with piezos. Since one of the possibilities for creating enough GDD to drive robust population transfer is by tuning the GTI in the laser cavity itself, we would ideally be able to get quasi-real-time readout of the pulse characteristics as we tweak the optics and the mode-locked laser, so the scan we desire should be fast and automatic. Furthermore, in order to characterize pulse chirp, we require both an intensity and electric field autocorrelator (either one by itself provides no information about pulse chirp). Of these, the electric field autocorrelator is more challenging, because it is a scanning Michelson interferometer, and fringe coherence must be maintained over the full throw of the delay. To our knowledge, commercial autocorrelators that can operate with these constraints not available, so we developed our own.

To produce an autocorrelator that works with pulses that are tens of picoseconds long requires a delay scan on the order of 1 cm. In order to do this without introducing vibrations that will interfere with the fringe coherence, we constructed a spinning block of high-index material that levitates on a homemade air bearing. The block of material is actually a container with two microscope slides for windows that we fill with water. This solution gave much cleaner interference fringes than we were able to obtain by polishing Lucite, and was less expensive than an optical flat of this size (many centimeters thick with a large area). As this delay block rotates, any laser beam passing through it is refracted through an amount of material that depends upon the angle of the block, and a retro-reflection on the other side using a corner cube sends the beam right back through the delay in a way that precisely cancels the displacement due to refraction. In this way, large angles can be used without displacing the beams as they re-combine on the beam splitter. The nonlinearity of the delay with angular position was canceled to first order by sending both arms of the interferometer through the same block at roughly right angles. The red trace in the top of Figure 3 shows a highly linear displacement with time achieved with this cancellation method.



**Figure 3. Performance of the long-pulse electric field autocorrelator. TOP:** Photodiode signal as a function of time for a mode-locked (blue) or CW (pink) laser input. The red trace shows the instantaneous fringe frequency, which is desired to be as flat as possible. **MIDDLE:** Horizontal zoom in of the mode-locked laser signal. **BOTTOM:** More horizontal zoom reveals high-quality Michelson interference fringes, which allows us to calibrate the time base to produce an accurate autocorrelation. This autocorrelator has a usable dynamic range of roughly 20,000 fringes at 778 nm.

Figure 3 shows the traces obtained with this autocorrelator. The usable range is revealed by sending a CW laser into the autocorrelator, which shows roughly flat, high-contrast interference fringes over a delay range of more than 50 ps. Zooming in on these traces (middle and bottom of Fig. 1) eventually shows the individual Michelson interference fringes. With our device, we can count more than 20,000 fringes at 778 nm, which demonstrates an auto-scanning, interferometrically stable delay range of 1.5 cm.



**Figure 4.** Fluorescence from cold Rb atoms after being excited by a single pulse from a mode-locked laser. The purple trace is experiment, the red trace is a convolution of a perfect 26.2 ns exponential decay with our detection system impulse response function.

We have also illuminated the CW MOT of rubidium atoms with single (transform-limited) pulses to quantify the excitation probability. Single pulses were selected from the mode-locked laser using an electro-optic pulse picker and focused onto a cloud of laser-cooled rubidium atoms. Figure 4 shows the fluorescence signature of excited Rb atoms, which decay from the  $^2\text{P}_{3/2}$  state with a time constant of 26.2 ns. The red curve is a theoretical calculation based on convolving an exponential decay with the Green's function of our detector. While this does not provide us with a benchmark for the excitation probability, it demonstrates single-pulse excitation, which is the first step toward substantial momentum transfer.

### Section III. Cryogenic Buffer-Gas Beam (CBGB)

Construction of a cryogenic buffer-gas beam (CBGB) source was also completed during the period of support from this program. The vacuum chamber has a square cross-section to provide large-area side plates that can be machined and fitted with custom capabilities such as viewports and connections to other vacuum system, and is shown in Fig. 5.

The creation of large vacuum chambers with a square cross section is a well-known challenge for those making CBGBs. Extruded tubing with square cross-section and thick walls is essentially not available in sizes over 10 inches on the sides, so groups relying on purchased tubing struggle with fitting everything into the chamber. On the other extreme is welded vacuum chambers, where the expense, probability of leaks, and bulk of the resulting chamber present drawbacks. We have pursued a new manufacturing method that we believe has solved many of these problems by creating our own thick-walled, square cross-section aluminum “tubing” with up to 14 inches on a side. The manufacturing was done by Electric Discharge Machining (EDM) of a single 14-inch cube of Al6061. A series of nested square vacuum chambers was cut from this one piece, and then sections were machined with dovetailed o-ring grooves and fixturing holes in the UCLA machine shop. In this way we were able to obtain the chamber bodies for at least three CBGBs from one manufacturing step.

The radiation shields, buffer-gas cell, buffer-gas feed line, and windows are all complete and working well. The cell currently reaches a temperature of about 5 K in a few hours after tuning on the



**Figure 5.** Completed CBGB. Shown in “closed cell mode” for spectroscopy. This system reaches a base temperature of about 5K.

system. We have not run the system in “beam mode,” but it has been transferred to another project and development continues there.

## Section IV. Two-photon laser cooling with an optical frequency comb

During our build-up of the system described in section II (the results of which are shown in Fig. 4), Andrew Jayich had demonstrated an optical frequency comb based MOT on a 1-photon transition. There were a couple of comb teeth used for this cooling and trapping, but most of the laser power was not involved in the cooling or trapping. We have since then been able to utilize 100% of the optical frequency comb’s laser power in Doppler cooling by driving a 2-photon transition. We stress here that we are far-detuned from the intermediate state, so this is a direct 2-photon transition (as opposed to stepwise 2-photon excitation). In this regime, a transform-limited optical frequency comb drives the transition at the same rate as a CW laser with the same time-averaged power, but with the added advantage that many other transitions within the comb bandwidth (repumps, other cooling lines, etc.) can also be driven with this strength without requiring a new laser or sacrificing power by modulation. The utility of this technique lies with species whose optical transitions lie deep in the UV, where very little CW laser power is available. Species sharing this constraint include H, C, N, and O, so the most prevalent atoms in chemistry may become available in ultracold form using this technique.

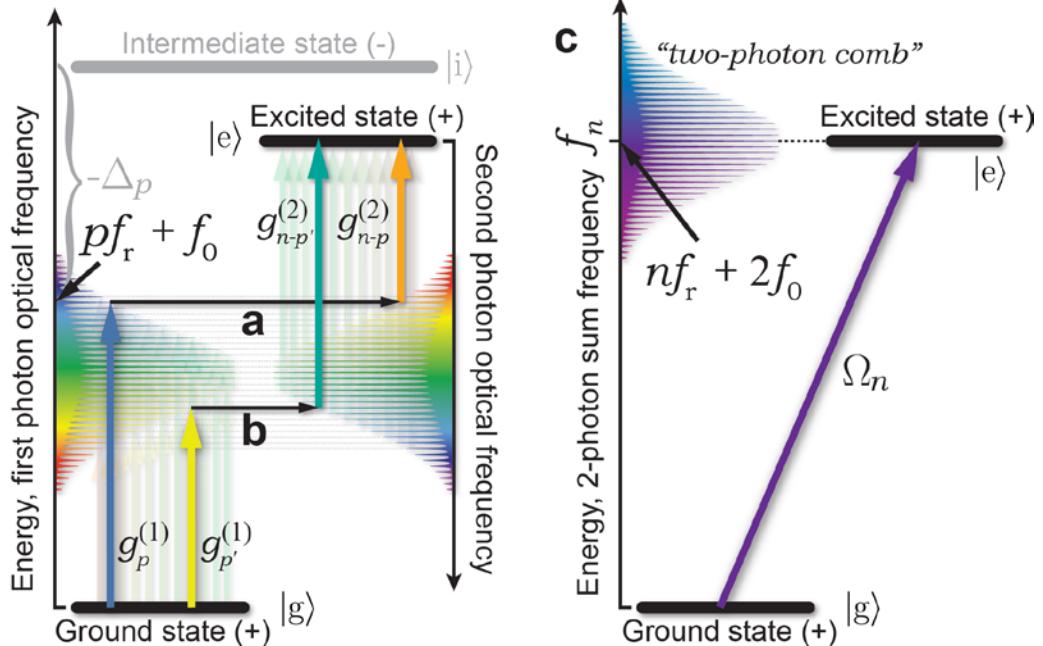
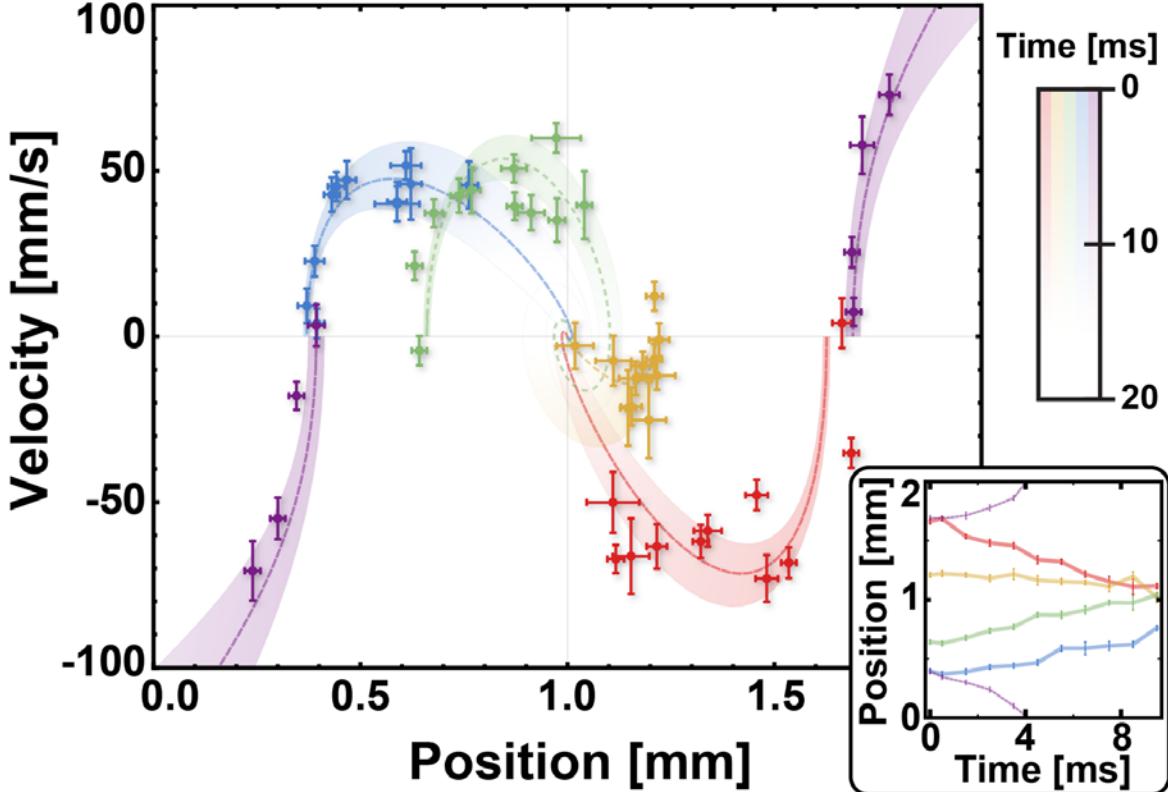


Figure 6. A transform-limited optical frequency comb can drive a 2-photon transition through massively-parallel constructive interference of all pathways (such as a and b, above) that satisfy conservation of energy. The resulting system resembles a 2-level system driven by a single, “two-photon comb” of coupling strength.

Figure 6 shows the concept by which an optical frequency comb can drive 2-photon transitions without wasting any optical power. All pairs of comb teeth whose sum frequency equals the transition frequency can contribute if the comb is transform-limited.



**Figure 7.** Laser cooling and trapping of atoms using an optical frequency comb. Phase-space trajectories are shown for atoms displaced from the MOT center by various amounts. Trajectories spiral in to the trap center, which demonstrates both confinement and dissipation. Purple traces show the anti-confining force for reversed polarization of the mode-locked MOT beams. The inset shows the same data in the time-domain as a slice through the position axis.

Figure 7 shows the behavior of rubidium atoms in a MOT formed by an optical frequency comb driving the 2-photon  $5D \leftrightarrow 5D$  transition. A CW MOT of Rb was loaded and then displaced from its equilibrium position. The CW beams were turned off and a 1D, counter-propagating mode-locked beam with proper polarizations for trapping was turned on. The resulting atomic motion demonstrates that this mode-locked MOT has a trap frequency of about 40 Hz and a damping frequency of about the same.

We argue in ref. [4] that this process can be used to trap atomic hydrogen on  $1S \leftrightarrow 3D$  at 243 nm. By choosing a rep rate carefully, all six of the allowed hyperfine and fine-structure transitions in this manifold can be driven simultaneously and close to resonance on the red side.

#### Section IV. Description of publications during period of support

The two most relevant publications that were enabled by the support of the Young Investigator Program are the theoretical paper we wrote in 2014 [1] and the latest paper from our group [4], which is under review at a journal. Though it was not what we set out to do (see below), ref. [4] in many ways represents the final product of this program, and support for continuation of that experiment has been moved to the NSF through their CAREER program. We also published two more theoretical papers with our colleagues at Georgetown University that focus on quantum simulation, which is a major application of cold molecules. A more-detailed look at these publications can be found below.

The work described in the proposal for this YIP grant formed the basis for a more-detailed theoretical analysis of the proposed method for creating samples of ultracold molecules. Starting with this program, we worked with colleagues from three other institutions to flesh out the scheme and simulate its performance, described in ref. [1]. Andrew Jayich, a postdoc in my group who was supported

solely by this grant, led this work and performed the simulations. Our conclusion was that the process looks to be capable of slowing and cooling a beam of molecules in about a 1 cm distance.

We also published a theory paper with collaborators that describes a type of spectroscopy for quantum many-body systems [2]. The spectroscopic techniques are very general, and as such have the potential to be applied to many platforms for quantum emulation, including cold molecules. In ref. [3], we describe the physics of self-assembled crystals of ions in Paul traps. This type of self-assembly is generically also expected for aligned electric dipoles (such as polar molecules), and the study of this in 2D may have relevance to cold, trapped molecules in the long-term future.

Last, ref. [4] reports the cooling and trapping of atoms using optical frequency combs. Even though the detailed experiments needed to publish this work were eventually supported by the NSF, the discovery of this process and the initial work on quantifying it were entirely due to the Young Investigator Program. Andrew Jayich, the postdoc supported by this YIP grant, deserves credit for discovering that the mode-locked laser we had been using to demonstrate stimulated forces could also make a MOT on its own. This would not have happened if we hadn't been trying to see the mechanical effects of a mode-locked laser on a MOT of atoms.

- [1] A. M. Jayich, A. C. Vutha, M. T. Hummon, J. V. Porto, and W. C. Campbell, *Phys. Rev. A* **89**, 023425 (2014)
- [2] Bryce Yoshimura, W. C. Campbell, and J. K. Freericks. *Phys. Rev. A* **90**, 062334 (2014)
- [3] Bryce Yoshimura, Marybeth Stork, Danilo Dadic, Wesley C. Campbell, and James K. Freericks. *EPJ Quantum Technology* **2**, 2 (2015)
- [4] A. M. Jayich, X. Long, and W. C. Campbell (submitted to PRX) arXiv: 1603.08053v1

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- [2] Bryce Yoshimura, W. C. Campbell, and J. K. Freericks. Phys. Rev. A 90, 062334 (2014)
- [3] Bryce Yoshimura, Marybeth Stork, Danilo Dadic, Wesley C. Campbell, and James K. Freericks. EPJ Quantum Technology 2, 2 (2015)
- [4] A. M. Jayich, X. Long, and W. C. Campbell (submitted to PRX) arXiv: 1603.08053v1

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